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CURRENT STATUS OF IRRADIATION TESTING OF THORIUM FUELS AT OAK RIDGE NATIONAL LABORATORY

A.R. Olsen, J.H. Coobs, and J.W. Ullmann

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CURRENT STATUS OF IRRADIATION TESTING OF THORIUM FUELS AT OAK RIDGE NATIONAL LABORATORY

A. R. Olsen, J. H. Coobs, and J. W. Ullmann

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CURRENT STATUS OF IRRADIATION TESTING OF THORIUM FUELS AT OAK RIDGE NATIONAL LABORATORY

A. R. Olsen, J. H. Coobs, and J. W. Ullmann*

ABSTRACT

The current status of irradiations involving principally sol-gel-derived pure and mixed compounds of thorium, uranium, and plutonium as oxides and carbides is discussed. In the bulk oxide irradiations the sol-gel material fabricated by vibratory compaction shows performance characteristics equivalent to pressed and sintered pellets at burnup levels in excess of 100,000 Mwd/tonne of heavy metal. In the coated particle tests the results indicate the need for a two-layer pyrolytic carbon coating and show that oxide fuel particles perform as well, or better than, carbide particles.

INTRODUCTION

Knowledge of the irradiation behavior of fuel materials is one of the basic requirements in the design of reactors for economical power production. Evaluation studies[1,2] of existing designs utilizing thorium indicate that only the molten salt converter and gas-cooled reactors currently are competitive with reactors fueled with natural or low-enrichment uranium fuels. Other studies[3,4] indicate that thorium-base fuels, using by-product plutonium from light water power reactors for fissile enrichment in a heavy water moderated reactor, may offer an economic means of establishing the thorium-233 uranium fuel cycle. The basic material characteristics[5] and the irradiation behavior of the thorium alloys and compounds are strongly dependent on the processing and fabrication techniques used. These characteristics must be defined to provide the designers with the maximum latitude in developing a specific economical design for any of the reactor concepts.

*Chemical Technology Division

The Oak Ridge National Laboratory sol-gel process offers a unique means of providing pure or mixed compounds of thorium, uranium, and plutonium as oxides and carbides.[6] These fuels have been combined with fabrication procedures such as vibratory compaction for metal-clad bulk oxide fuels or coating procedures utilizing pyrolytic carbon as a fission-product retention material with and without subsequent incorporation into fueled graphite loadings. Thus, the full range of processing and fabrication techniques are incorporated into a material for irradiation evaluation. The irradiation behavior of solid thorium-base fuels has been under investigation at ORNL for several years. Most of the data have been reported as it developed.[7,8] This paper will summarize only our findings and establish the current status of the programs. The irradiation testing can be subdivided into the two basic fuel types.

Metal-Clad Bulk Oxides

A series of noninstrumented fuel rods have been irradiated in the process water of the Materials Test Reactor, Engineering Test Reactor, and the Chalk River National Research Experiment Reactor during the past six years in the metal-clad bulk oxide program. Two more sophisticated test groups, one with instrumented capsules and one loop bundle, were irradiated in the Oak Ridge Research Reactor. The program has been concentrated on the sol-gel-derived fuels with low fissile content (approximately 5% UO_2 in ThO_2) fabricated into fuel rods by vibratory compaction.

Table 1 is a listing of the irradiation tests. While most of the original irradiation tests utilized stainless steel cladding, the more recent rods have been fabricated using Zircaloy-2 cladding materials and the tests have been designed to evaluate the effects of semiremote fabrication and different sol-gel calcination atmospheres. Pure thoria without any initial fissile additive is being irradiated in the most recent tests to investigate the effects of increasing power level.

Table 1. Summary of Thorium Fuel Cycle Program Irradiation of Powder-Packed Rods[a]

Designation	Number of Rods	Type of Oxide	Density (Percent Theoretical)	Linear Heat Rating (w/cm)	Peak Burnup (Mwd/tonne metal)	Objective
MTR-I	7	Arc-fused, sol-gel E	86 to 87	390	15,000–100,000	Provide base-line data to use in comparing sol-gel and arc-fused oxide
MTR-II[b]	2	Sol-gel S	88 to 89	600	100,000	Obtain higher heat rating by increasing enrichment
MTR-III[b]	6	Sol-gel 35	86 to 89	820	100,000	Compare oxide calcining atmospheres and higher heat ratings obtained by increasing diameter
ETR-I	4	Sol-gel 35	86 to 89	960	22,000	Same as for MTR-III
NRX-I	8	Sol-gel A and B	86 to 87	160	16,000	Provide base-line data
NRX-II	4	Sol-gel C, arc-fused	83 to 86	210	5,000	Study effect of increased length
NRX-III	6	Sol-gel S	88 to 89	270	23,000	Study effect of increased length
NRX-III	3	Sol-gel ThO_2-PuO_2	74 to 76[c]	260	22,000	Study ThO_2-PuO_2 oxide and lower packed density
ORR Loop	3	Sol-gel 26	84 to 85	500	2,100	Study in pressurized water at 260°C and 1750 psi
ORR Poolside	2	Sol-gel D	85[c]	340	5,000	Measure effective thermal conductivity using a central thermocouple in NaK at 315 psi, 540 and 705°C
ETR-II[b]	6	BNL-sol-gel	90	630	30,000–100,000	Study effects of remote fabrication and oxide recalcining
ETR-III[b]	7	Sol-gel ThO_2	88	770	10,000–70,000	Study ThO_2 blanket material with gradually increasing heat rating and provide high Pa low-fission-product material for chemical processing

[a]All rods were clad with type 304 stainless steel, except for Groups ETR-II, -III, and ORR Loop, which were clad with Zircaloy-2.

[b]Currently under irradiation.

[c]Tamp-packed; all others vibratorily compacted.

The initial objectives of the irradiation program were to compare the characteristics of the chemically produced sol-gel $(ThU)O_2$ fuel with those of arc-fused material and to compare the performance of vibratorily compacted rods with others containing pressed and sintered pellets. The results of these early comparison tests have been favorable. In Table 2, for example, we have listed a group of tests all of which were conducted at moderate linear heat ratings between 300 and 460 w/cm (10 to 14 kw/ft), which is similar to current water reactor elements using UO_2 as a fuel. These tests were taken to high burnups where as much as 70% of the fissions occurred in the bred ^{233}U. Even with these burnups, in excess of 100,000 Mwd/tonne of heavy metal, the fission-gas release rates were generally less than 20%. Estimates of the swelling of the pellet fuels[9] based upon the fact that there was no change in diameter or length of the self-supporting claddings at the high burnup levels indicate the change in volume was less than 0.5% per 10^{20} fissions/cm^3. The postirradiation examination of vibratorily compacted and pellet fuels showed essentially no difference in the macroscopic appearance after extended irradiations. By comparing Fig. 1 with Fig. 2, where typical cross sections of a pellet rod and a sol-gel vibratorily compacted powder rod are shown, it can be seen that without the titles there is no easy way of separating the initial starting fuel form. Both rods show typical thermally fragmented fuel.

The current irradiation program is investigating the effects of process parameters such as semiremote fabrication or the sol-gel calcining atmosphere, and at the same time attempting to establish the maximum fuel performance characteristics. In one group of tests in this series, we have succeeded in developing central voids but with no evidence of melting. Figure 3 shows typical macroscopic sections similar to those of the lower heat rated rods but showing the central voids. Figure 4 is a compilation of radial micrographs from three of the highly rated rods. The rods operated at peak heat ratings in excess of 1000 w/cm, while the time-averaged heat ratings are those shown under each micrograph. In these highly rated rods less than 30% of the fission gas was released from fuels fired in nitrogen or in the standard reducing atmosphere of

Table 2. Comparative Operating Conditions for Various ThO_2-UO_2 Fuels

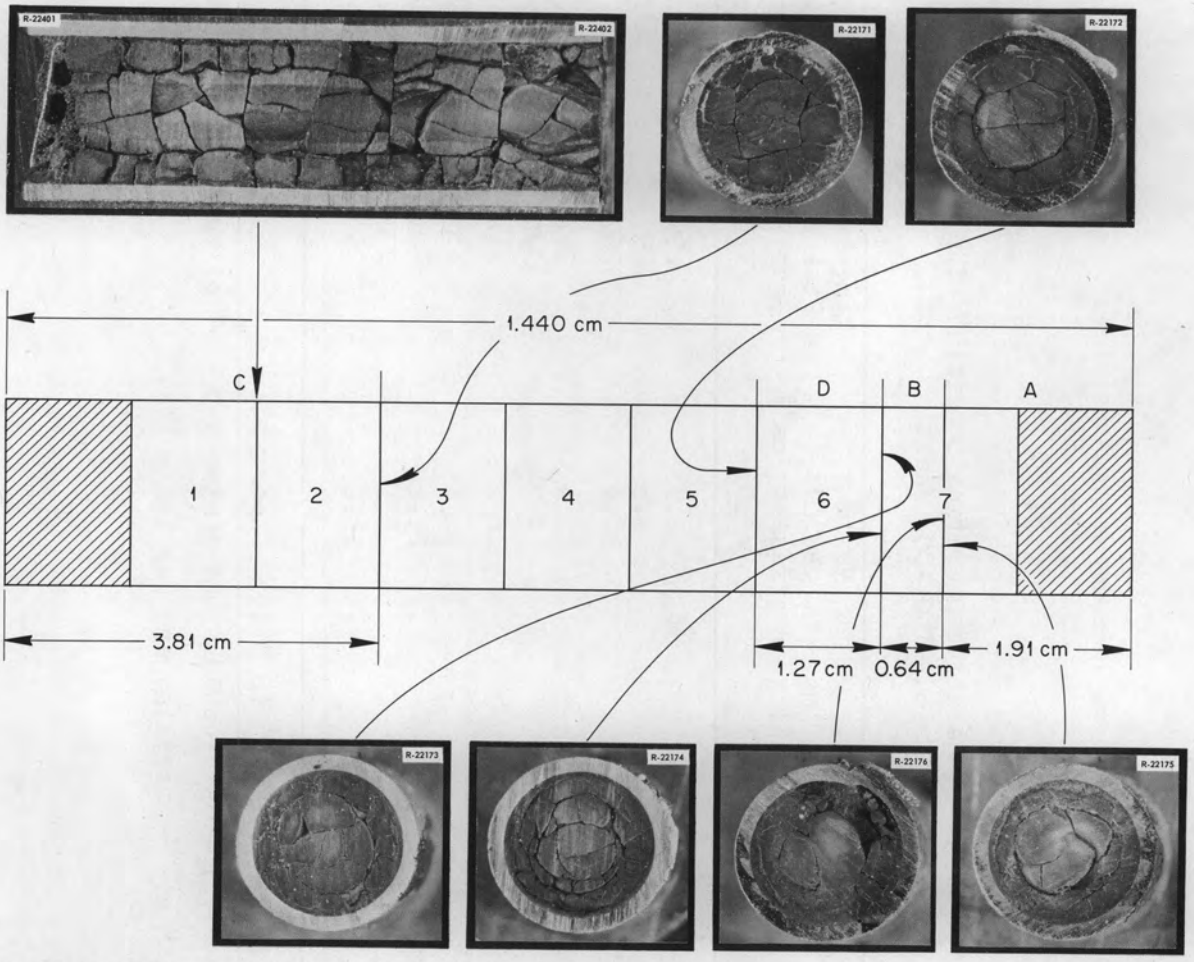
Fuel	Fuel Form	Irradiation Time (Reactor Full Power Days)	Maximum Burnup (fissions/cm^3) $\times 10^{20}$	Time-Averaged Peak Linear Heat Rating (w/cm)	$\int_{T_o}^{T_c} k d\theta$ (w/cm)	^{85}Kr Release (%)
Arc-fused ThO_2–4.5% UO_2	ViC[a]	110	2.5	299	33.8	2.4
	ViC	376	8.6	297	33.7	7.2
	ViC	707	14.4	267	31.6	6.4
Sol-gel ThO_2–4.5% UO_2	ViC	110	2.9	341	37.8	0.5
	ViC	375	8.2	286	32.8	13.2
	ViC	691	16.5	311	34.6	17.0
Pressed and sintered ThO_2–4.5% UO_2	Pellet[b]	905	26.4	420	44.8	22.8
	Pellet	660	21.1	461	48.2	12.4
	Pellet	497	8.1	270	31.5	c
	Pellet	406	11.0	394	42.6	c

[a]Vibratorily compacted to a density 85 to 87% of theoretical.

[b]Pellets pressed from coprecipitated powders and sintered to 93% of theoretical density.

[c]Gas samples diluted with air in sampling and partially lost.

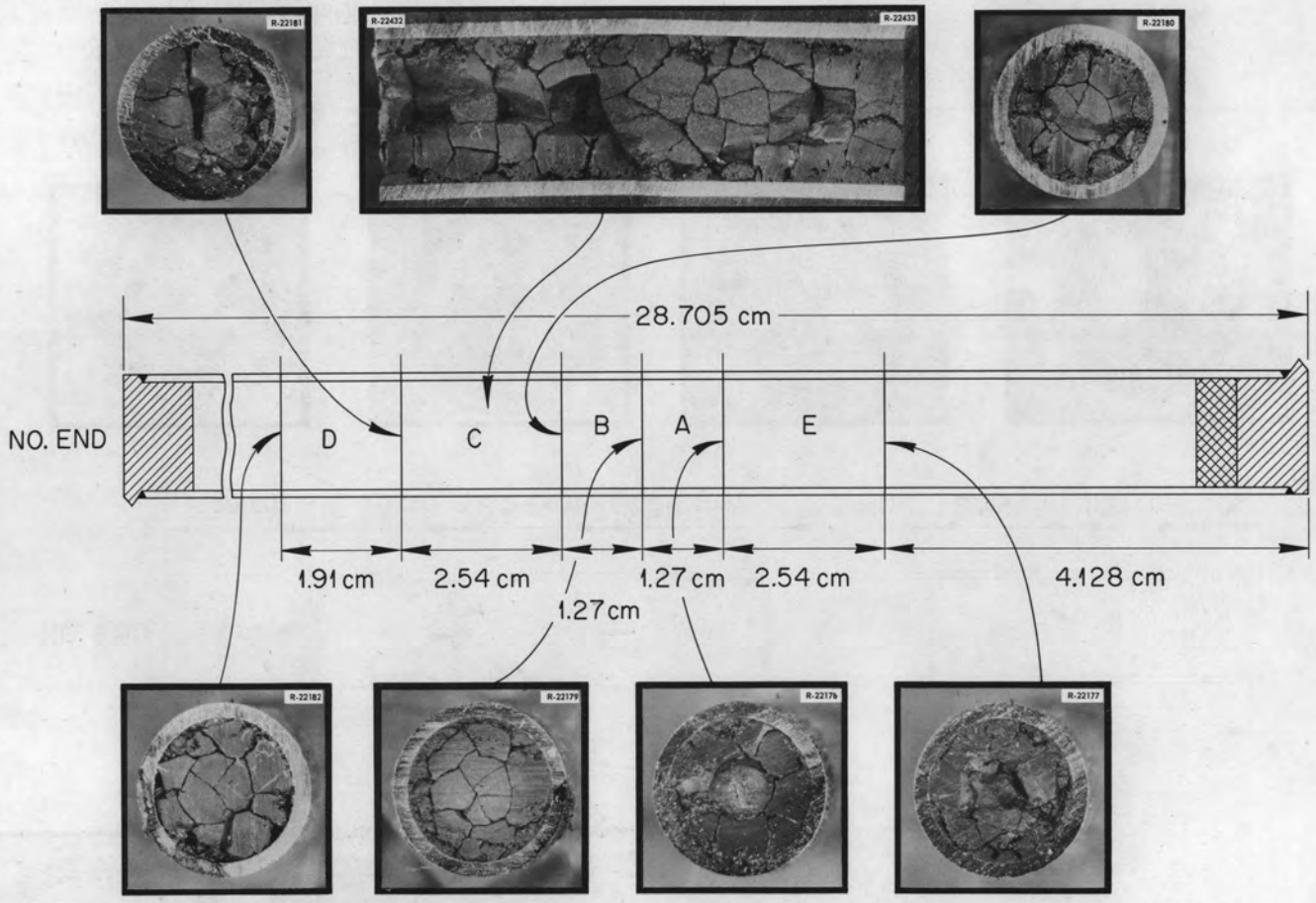
ORNL-DWG 65-2476



A. METALLOGRAPHY SECTION
B. BURNUP SECTION
C. LONGITUDINAL SECTION
D. NEUTRON ACTIVATION SPECIMEN

Fig. 1. Macroscopic Views of Selected Section Locations of Pellet Rod 712, as Cut.

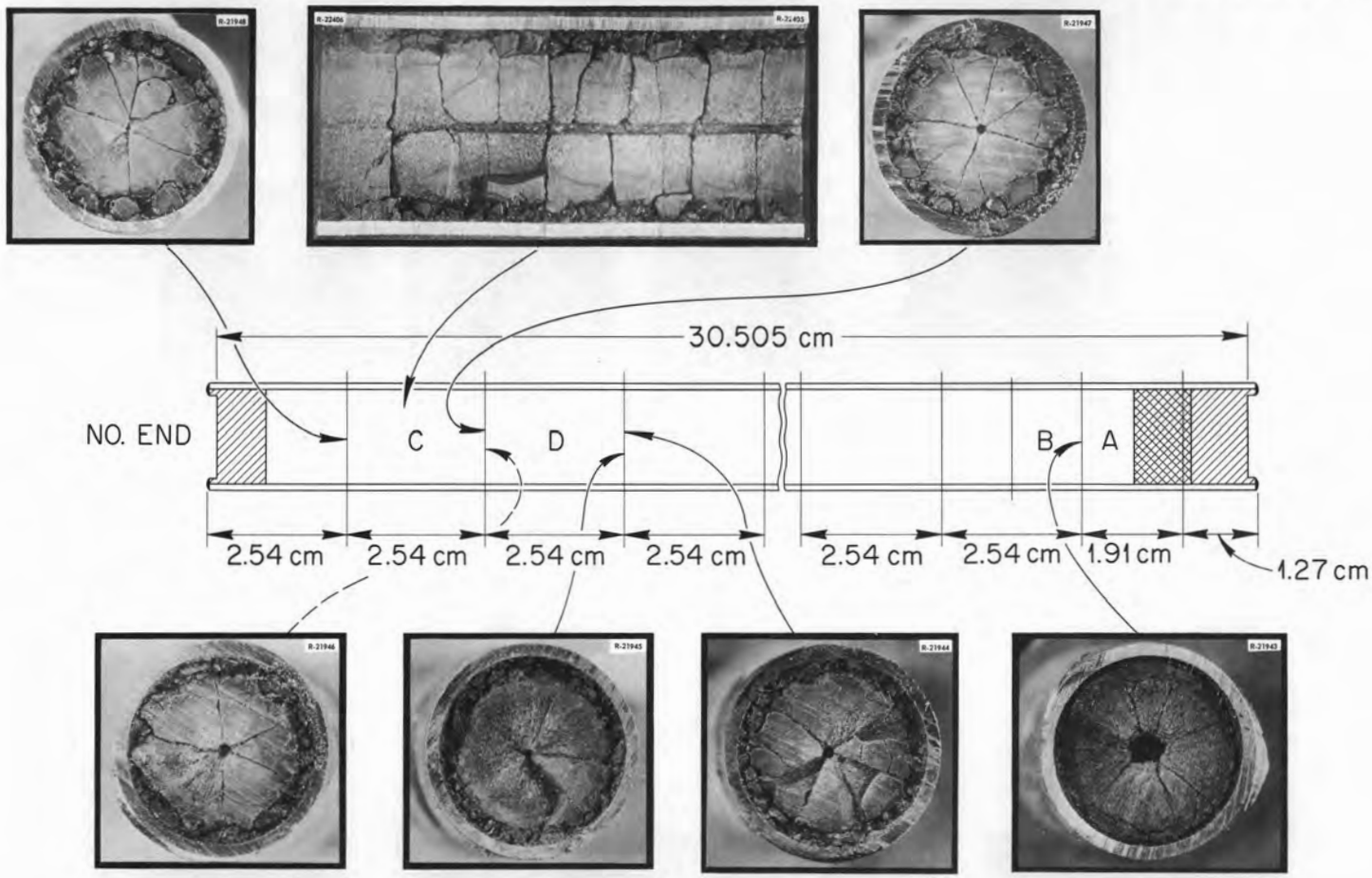
ORNL-DWG 65-2480



A. METALLOGRAPHY SECTION
B. BURNUP SECTION
C. LONGITUDINAL SECTION
D. SPARE LONGITUDINAL SECTION
E. NEUTRON ACTIVATION SECTION

Fig. 2. Vibratorily Compacted Sol-Gel Fuel, MTR Group I, Capsule Z-8, Macroscopic Views of Selected Section Locations, as Cut.

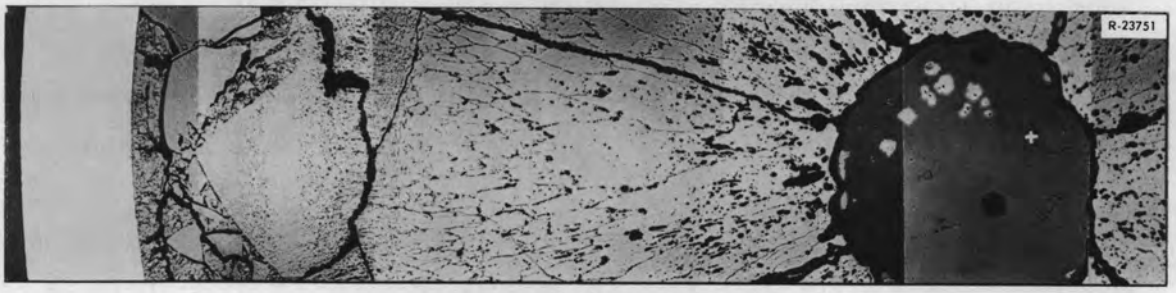
ORNL-DWG 65-2470



A. METALLOGRAPHY SECTION
B. BURNUP SECTION
C. LONGITUDINAL SECTION
D. SPARE LONGITUDINAL SECTION

Fig. 3. ETR Group I, Capsule 8, Macroscopic Views of Selected Section Locations, as Cut.

PHOTO 80962

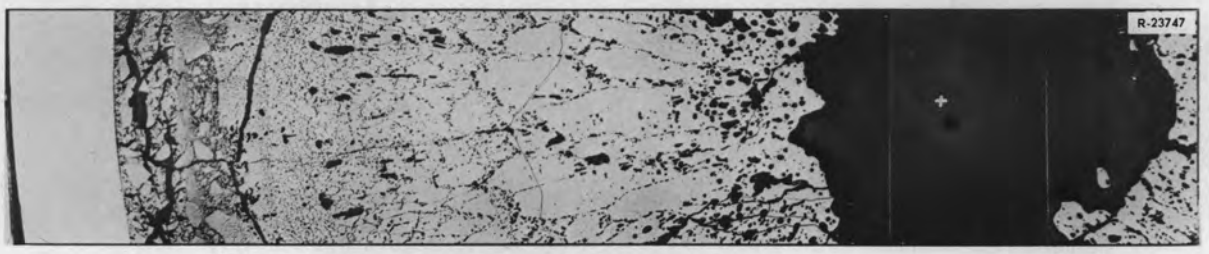


2.54 mm

AIR FIRED FUEL IRRADIATED TO 4.3×10^{20} fissions/cm^3 AT A LINEAR HEAT RATING OF 870 w/cm.

2.54 mm

FUEL FIRED IN Ar-4% H_2 AND IRRADIATED TO 4.3×10^{20} fissions/cm^3 AT A LINEAR HEAT RATING OF 865 w/cm.



2.54 mm

NITROGEN FIRED FUEL IRRADIATED TO 4.5×10^{20} fissions/cm^3 AT A LINEAR HEAT RATING OF 910 w/cm.

Fig. 4. Composite Radial Micrographs at Maximum Burnup Locations of Vibratorily Compacted Sol-Gel ThO_2–6% UO_2 Fuel Clad in 304L Stainless Steel.

Ar–4% H, while approximately 40% was released from the fuels calcined in air. The burnup level on these rods was 22,000 Mwd/tonne (Th+U). There was no evidence of swelling as determined by postirradiation measurements of the free-standing cladding. The apparent effect of the firing atmosphere on increased fission-gas release and microstructural changes will not be resolved until other tests using identical fuels but at lower linear heat ratings are examined. These tests are currently under irradiation and are scheduled to go to approximately 100,000 Mwd/tonne of heavy metal burnup.

Since only recently has a sol-gel process for PuO_2 been developed, our irradiation experience with the so-called mixed progeny fuels is limited. Three rods containing sol-gel-derived ThO_2 mixed, before calcining with precipitated 5.2% PuO_2, were tamp-packed to approximately 75% of theoretical density and exposed at a linear heat rating of 200 to 245 w/cm for a peak burnup of 29,000 Mwd/tonne (Th+Pu). The fission-gas release rates were less than 5% and the microstructures as shown in Fig. 5 were similar to those of ThO_2–5% UO_2 exposed under the same conditions. The center temperatures estimated by using a thermal conductivity of 0.017 w/cm, which was the effective thermal conductivity derived in the instrumented capsule experiments, were approximately 1500°C in rod P-4 and 1300°C in rod P-6. There is some incipient sintering and grain growth in the fuel at the higher heat rating while with rod P-6, except for accentuated distributed porosity in the center, the fuel is essentially unchanged from the preirradiation appearance.

Although the data on ThO_2-base fuels are still somewhat limited, a preliminary comparison with UO_2 fuels is in order. The data presented in Table 3 compare powder-compacted UO_2 and ThO_2-UO_2 fuels using the $\int k d\theta$ values for specific structural changes most commonly used in comparing bulk oxide fuels. The UO_2 work was done as part of the Maritime Reactor Program and was reported previously.[10] The ETR-I data were included to provide a value for the $\int k d\theta$ for void formation in the thoria-base fuels even though the surface temperature of the cladding was 150°C lower. The loop experiments were run in the same facility. There are uncertainties in the peak-to-average ratios of

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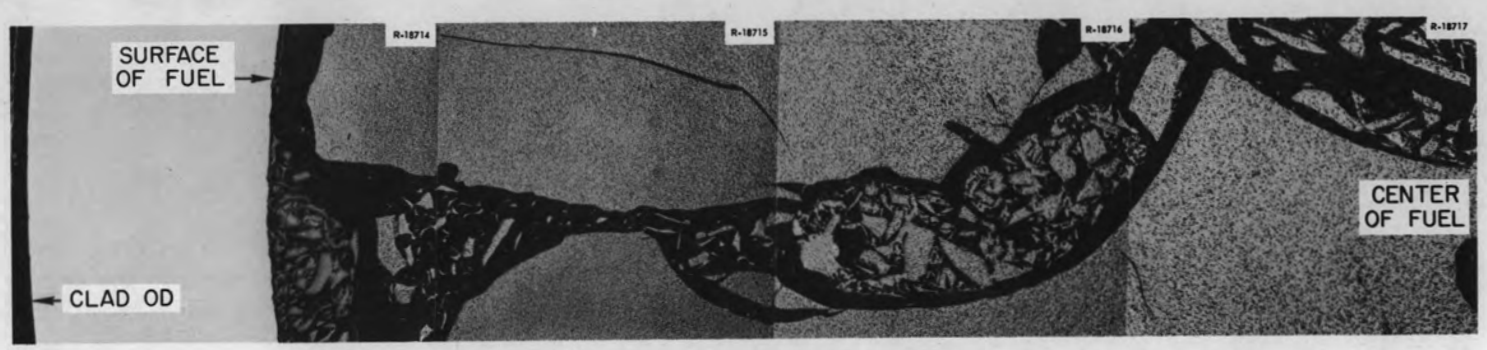
P-4 IRRADIATED (5.2×10^{20} fissions/cm^3) 100 X



P-6 IRRADIATED (4.65×10^{20} fissions cm^3) 100 X

Fig. 5. Composite Photomicrographs Showing Typical Transverse Sections of Irradiated Tamp-Packed Sol-Gel ThO_2–5.2% PuO_2. Radial area from center to clad surface. 100X.

Table 3. Comparison of UO_2 and ThO_2-UO_2 Vibratorily Compacted Fuel Rods

Experiment	Fuel Material[a]	Fuel Density (% TD)	Burnup Mwd/tonne Heavy Element	Inside Surface Temperature of Cladding (°C)	Linear Heat Rating (w/cm)	$\int_s^c k d\theta$[b] (w/cm)	$\int_s^v k d\theta$[b] (w/cm)	$\int_s^{cg} k d\theta$[b] (w/cm)	$\int_s^{eg} k d\theta$[b] (w/cm)	^{85}Kr Release (%)
ORR Loop	UO_2									
7N1		87.1	4,780	353	434		32.5	20.7		12.4
7O1		87.1	5,140	361	465		34.8	21.1		47.0
7P1		86.9	6,290	383	566		41.2	20.0		72.0
8N1		85.6	4,810	347	404		30.9	22.8	17.5	77.0
8O1		85.8	5,040	351	423		31.3	21.2	16.1	25.0
8P1		85.5	6,880	387	578		40.8	23.9	20.3	25.0
ORR Loop	Th–6% UO_2									
L1A		85.2	1,600	341	381	30.3	no void		20.5	2.3
L1B		84.1	2,100	368	499	39.7	no void	36.6	17.7	
L1C		84.1	1,730	348	410	32.6	no void		20.7	3.9
ETR-1	ThO_2–5% UO_2									
18		88.1	20,400	211	870		63.0	35.8	34.7	38.0
10		89.4	20,000	209	865		66.4	24.6	19.8	28.0
8		85.1	22,000	218	914		61.3	28.2	25.3	21.0

[a] UO_2 fuel was arc-fused crushed and vibratorily compacted; oxygen-to-uranium ratio 2.002:2.003; enriched in ^{235}U 5 to 6%. ThO_2-UO_2 fuel was sol-gel material crushed and vibratorily compacted; uranium enriched in ^{235}U to 93%.

[b] $\int_s^c$ is center to surface of fuel; $\int_s^v$ is void to surface of fuel; $\int_s^{cg}$ is the limit of columnar grain growth to surface of fuel; and $\int_s^{cg}$ is the limit of discernible equiaxed grain growth to surface of fuel.

the heat flux in these experiments. Nevertheless, a comparison of the heat ratings to produce similar microstructural changes and fission-gas release rates in the vibratorily compacted thoria-base fuels and the urania fuels indicates the thoria fuels can accommodate approximately 40% higher heat ratings.

Future tests are planned to investigate the sol-gel-derived $(Th\text{-}Pu)O_2$ fuels with emphasis on low-energy vibrational compaction of microspheres. In addition, tests utilizing thin-walled claddings which are not free-standing are planned to investigate the swelling characteristics of thoria-base fuels.

Coated Particles

The irradiation program on coated particle fuels is of more recent origin than the program on bulk oxides. The fuel has all been in the form of small particles (150–350-μ diam) of $(ThU)C_2$ and $(ThU)O_2$ coated with pyrolytic carbon for fission-product retention. Both loose beds of coated particles and fueled-graphite elements containing coated particles have been irradiated.[8] Much of the irradiation testing has been designed to evaluate the effects of changing coating parameters on the particle behavior as discussed by Prados et al. in another paper at this symposium.[11] In order to obtain accelerated burnup, high concentrations of uranium have been used for most of the test with this fuel form. The dicarbide particles have generally been obtained from the carbon bed melting process, while the oxides are from conventional sintering processes or the sol-gel microsphere process.

Table 4 is a partial listing of the tests utilizing fuel kernels containing thorium dicarbide. Some properties of these coated particles are given in Table 5. There were no observed coating failures in any of these tests. As can be seen from Table 4, the irradiation test temperature has varied from approximately 1000°C up to a maximum of 1500°C and the heavy metal fissioning has been carried to as high as 27 at. % for the low thorium-to-uranium ratio particles. Testing of the higher thorium-to-uranium ratios involves longer periods of exposure and only recently have coated fuel particles with the higher

Table 4. Irradiation Test Conditions and Fission-Gas Release Data for Coated $(ThU)C_2$ Particles

Sample Designation	Thorium-to-Uranium Ratio	Coating Structure	Test Temperature (°C)	Burnup (at. %) Heavy Metal Fissioned	R/B for ^{88}Kr
NCC-214	0.6	Duplex	1530[a]	2.4	2×10^{-4}
NCC-208	0.6	Duplex	1175	1.7	1.6×10^{-5}
NCC-208	0.6	Duplex	1125	6.0	1.3×10^{-6}
NCC-208	0.6	Duplex	1300	11.	b
NCC-208	0.6	Duplex	1500	12.	b
NCC-210	0.6	Duplex	~ 1000[a]	27.	b
GA-310	0.6	Triplex	1125	10.	3.6×10^{-6}
GA-N-1736-121E	0.6	Triplex	~ 1000[a]	27.	b
GA-314	2.2	Triplex	1400	0.3	2.6×10^{-5}
GA-314	2.2	Triplex	1400	8.	3.5×10^{-5}
NCC-222	4.9	Duplex	1300	10.	3.8×10^{-5}

[a]Irradiated in fueled-graphite spheres; all others irradiated as unsupported.

[b]Not measured; static experiment.

Table 5. General Information on Pyrolytic-Carbon-Coated Particles Used in Irradiation Tests

Particle Batch Number	NCC-208	NCC-210	NCC-214	GA-310	GA-314	N-1736-121ECB
Core material	$(UTh)C_2$	$(UTh)C_2$	$(UTh)C_2$	$(UTh)C_2$	$(UTh)C_2$	$(UTh)C_2$
Uranium content, wt %	19.6	23.4	23.0	14.4	18.9	19.9
Thorium content, wt %	13.4	13.6	13.4	9.0	38.7	12.2
Enrichment, at. %	93	93	93	93	93	92.9
Particle dimensions, μ						
Total particle, av	445	432	424	470	550	426
Core particle, av	213	216	218	175	349	200
Coating thickness, av	117	108	103	148	101	113
Coating structure	duplex	duplex	duplex	triplex	duplex	triplex
Pour density, g/cm^3	1.6	1.8	1.7	1.5	2.2	a
Particle weight, av ($g \times 10^{-4}$)	1.2	1.2	1.2	1.4	3.5	a
Coated-particle density,[b] g/cm^3	2.79	2.95	2.80	2.37	3.58	2.70
Crushing load, av (g)	1473	1320	1456	1598	1531	1290
Surface contamination, $\%U \times 10^{-3}$	1.8	3.9	2.0	2.4	7.9	a
Fuel removal by leaching, $\%U \times 10^{-3}$	< 0.5	0.5	17.0	0.2	0.7	a

Table 5 (continued)

Particle Batch Number	NCC-222	HB-23	OR-182	OR-205	OR-206
Core material	$(ThU)C_2$	UO_2	$(ThU)O_2$	ThO_2	$(ThU)O_2$
Uranium content, wt %	5.55	17.8	3.3		3.1
Thorium content, wt %	25.4		39.8	33.8	36.8
Enrichment, at. %	93	97	93		93
Particle dimensions, μ					
Total particle, av	437	407	410	500	406
Core particle, av	214	148	217	243	206
Coating thickness, av	112	129	96	128	100
Coating structure	duplex	triplex	duplex	duplex	duplex
Pour density, g/cm^3	1.66	1.54	1.7	1.7	1.8
Particle weight, av ($g \times 10^{-4}$)	1.15	0.82	1.1	a	1.3
Coated-particle density,[b] g/cm^3	2.67	2.43	a	2.87	2.94
Crushing load, av (g)	1780	1210	1270	3210	2730
Surface contamination, $\%U \times 10^{-3}$	8.7	0.16	20.2	3.1	6.7
Fuel removal by leaching, $\%U \times 10^{-3}$	2.3	< 0.01	275	0.3	2.4

[a]Not measured.

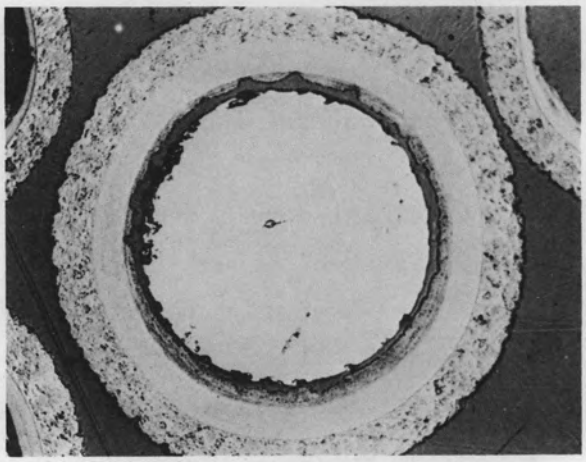
[b]Measured with helium pycnometer.

thorium-to-uranium ratios of 2.2 and 4.9 been tested at significant levels of fissioning. Preirradiation and postirradiation micrographs of batch GA-314, which has a thorium-to-uranium ratio of 2.2:1, are shown in Fig. 6. After 8 at. % heavy metal fissioning the two outer layers of the triplex coating appear unchanged, indicating that the porous inner coating has absorbed all direct fission recoil damage and also accommodated any swelling of the fuel particle. Such good behavior probably would be predicted by the mathematical model discussed by Prados[11] if detailed properties of the individual layers of the coating were well documented.

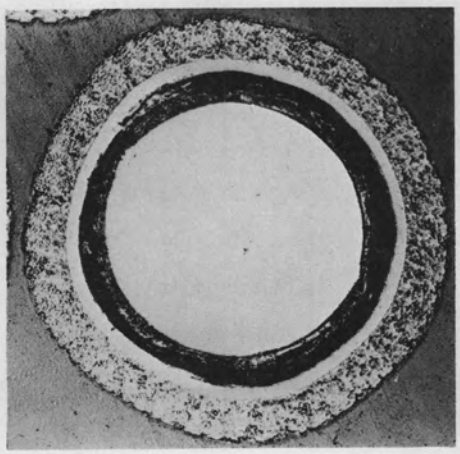
Perhaps the most significant development in the area of coated fuels during the past year has been the excellent performance of pyrolytic-carbon coated sol-gel-derived oxide microspheres. The deposition conditions used for coating sol-gel oxide microspheres are given in Table 6, while properties of the coated particles were presented in Table 5. Table 7 is a listing of the irradiation tests on these fuels which have been completed to date. The thorium-to-uranium ratio in the mixed oxide particles has been 12:1 and, consequently, the fuel burnup has been restricted. However, the test of fully enriched UO_2 sol-gel microspheres coated with a thick three-layer coating, which was exposed at 1600°C to a burnup of 25 at. % heavy metal fissioned, clearly shows the excellent performance of the oxide particles. Again there were no observed failures in any of these tests.

Of greater importance than the lack of any failed particles is the fact that the fission-gas release rates, as defined by the R/B ratios, are as low or slightly lower than those from the coated-carbide particles shown in Table 4. We attribute the lower release rates to the reduced coating contamination normally observed with carbon-coated oxide particles. Contamination of the coatings by fuel usually occurs during deposition of the high-density outer coating at high temperatures ($\geq$ 1800°C). The fission-gas release rates from many previous experiments have indicated a correlation with coating contamination. In a recent series of sweep-capsule irradiations using both coated UO_2 and $(ThU)C_2$ particles, we were able to verify that the observed release is

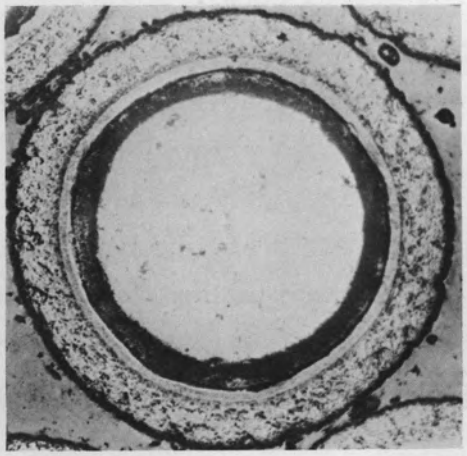
R-28857



UNIRRADIATED



1400°C 0.3% B.U.
HVY. METAL



1400°C 8% B.U.
HVY. METAL

Fig. 6. Fuel Batch GA-314, Triplex-Coated 350μ Diameter $(Th,U)C_2$, 2.2:1. Average total coating thickness 101μ. No particles failed.

Table 6. Deposition Conditions of Coatings on Sol-Gel Oxide Microspheres

	Sample Designation			
	OR-182	OR-205	OR-206	HB-23
Type of fuel particle	$(ThU)O_2$[a]	ThO_2	$(ThU)O_2$[a]	UO_2
Average fuel particle diameter, μ	217	243	206	148
First layer				
Coating temperature, °C	1400	1400	1400	980–1050
CH_4 flow rate, $cm^3\ min^{-1}\ cm^{-2}$	0.15	0.83	0.83	2.3[b]
Coating thickness, μ	54	50	35	49
Second Layer				
Coating temperature, °C	1800	1800	1800	1320
CH_4 flow rate, $cm^3\ min^{-1}\ cm^{-2}$	0.33	0.17	0.17	1.20
Coating thickness, μ	42	78	65	20
Third Layer				
Coating temperature, °C				1900
CH_4 flow rate, $cm^3\ min^{-1}\ cm^{-2}$				0.05
Coating thickness, μ				60

[a]Particles contain 8 wt % UO_2, highly enriched.

[b]Acetylene was used for inner coating on HB-23.

Table 7. Irradiation Test Conditions and Fission-Gas Release Data for Unsupported Coated Sol-Gel Oxide Microspheres

Sample Designation	Type of Fuel Particle[a]	Coating Structure	Test Temperature (°C)	Burnup (at. %) Heavy Metal Fissioned	R/B for ^{88}Kr
OR-206	$(ThU)O_2$	Duplex	1200	0.6	2.3×10^{-6}
OR-206	$(ThU)O_2$	Duplex	1370	2.7	2.5×10^{-5}
OR-205	ThO_2	Duplex	1370		
OR-182	$(ThU)O_2$	Duplex	1200	0.25	b
HB-23	UO_2	Triplex	1600	25	4.0×10^{-8}

[a]Thorium-to-uranium ratio in all mixed fuels equals 12:1.

[b]Not measured; static experiment.

a direct function of the amount of fissionable material in the coatings, as shown in Fig. 7. Such a correlation applies only if no particles are broken.

The thermal stability of pyrolytic-carbon coated oxides is remarkably good at temperatures much higher than the irradiation tests listed in Table 7. An extensive series of heat treatments on coated ThO_2 and UO_2 particles at 1900 and 2000°C demonstrated that high-density outer coatings successfully contain CO formed by reaction of the oxides with carbon.[12] Since the reaction is thus inhibited, little or no carbide is formed and migration of fuel into the coating is virtually undetectable. The striking difference in fuel migration behavior of UO_2 and UC_2 with similar coatings is illustrated in Fig. 8. Both types of particles have porous inner coatings (density < 1.0 g/cm^3) deposited from acetylene.

The obvious process advantage of being able to handle the uncoated particles without the use of an inert atmosphere, when combined with the lower fuel swelling of the oxide, indicates a potential advantage for these fuels. The basis for predicting reduced swelling as developed from the examination of the bulk oxide fuel has been discussed elsewhere.[13] This same discussion indicates an advantage for thorium-containing fuels over those containing only fissile uranium. These fuel characteristics together with the variety of coating parameters have been included in the model discussed in the paper by Prados _et al._, which describes the prediction of coated-particle performance. A series of noninstrumented capsule irradiations was recently initiated to test the model. The results of postirradiation examination of the first capsule in the series have been included in that paper. A later experiment in the series is specifically designed to investigate the effects of the thorium-to-uranium ratio on fuel particle behavior.

To date, none of our irradiation tests have shown any significant difference in the performance of dicarbide or oxide fuel particles regardless of thorium content. As we have seen in Fig. 6 and as listed in Table 4, there are $(ThU)C_2$ particles with both two- and three-layer coatings that have shown good performance to high burnup levels. A

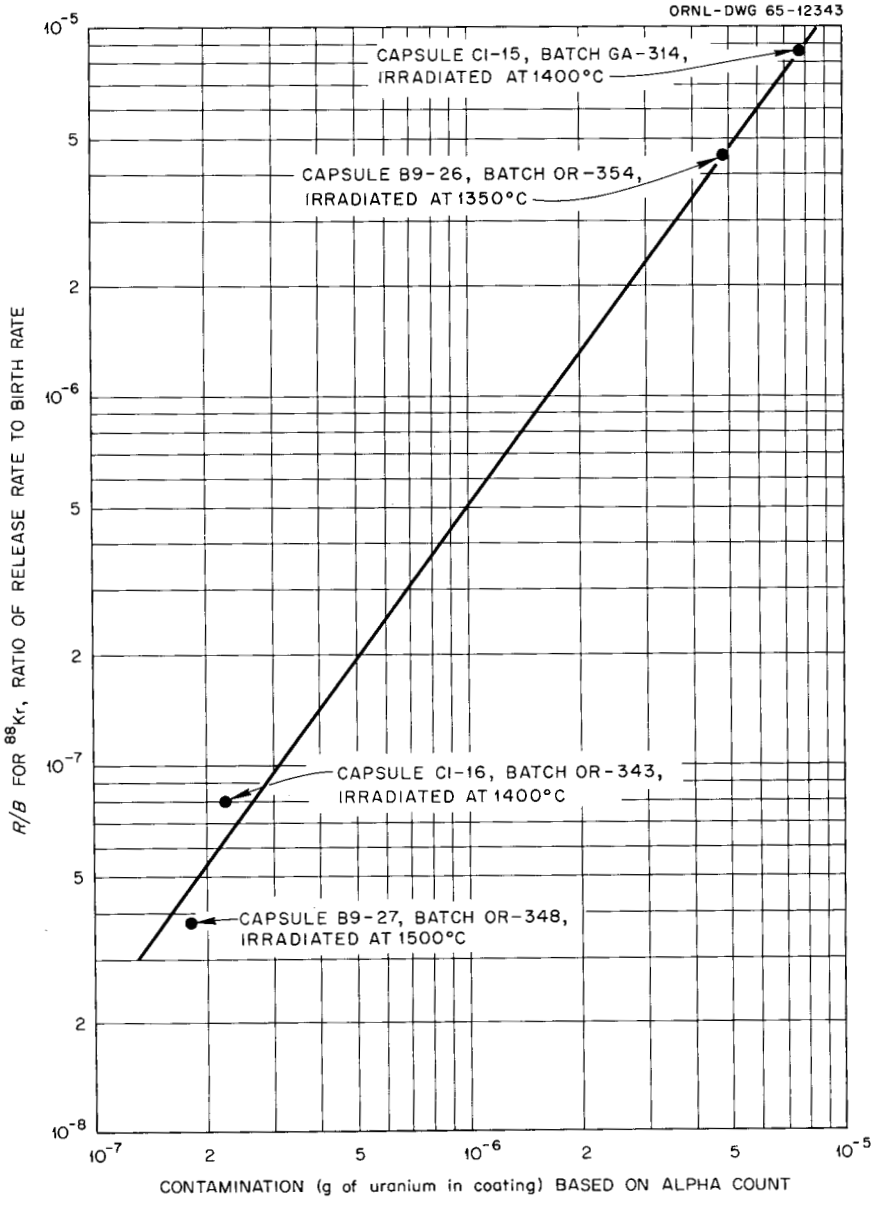
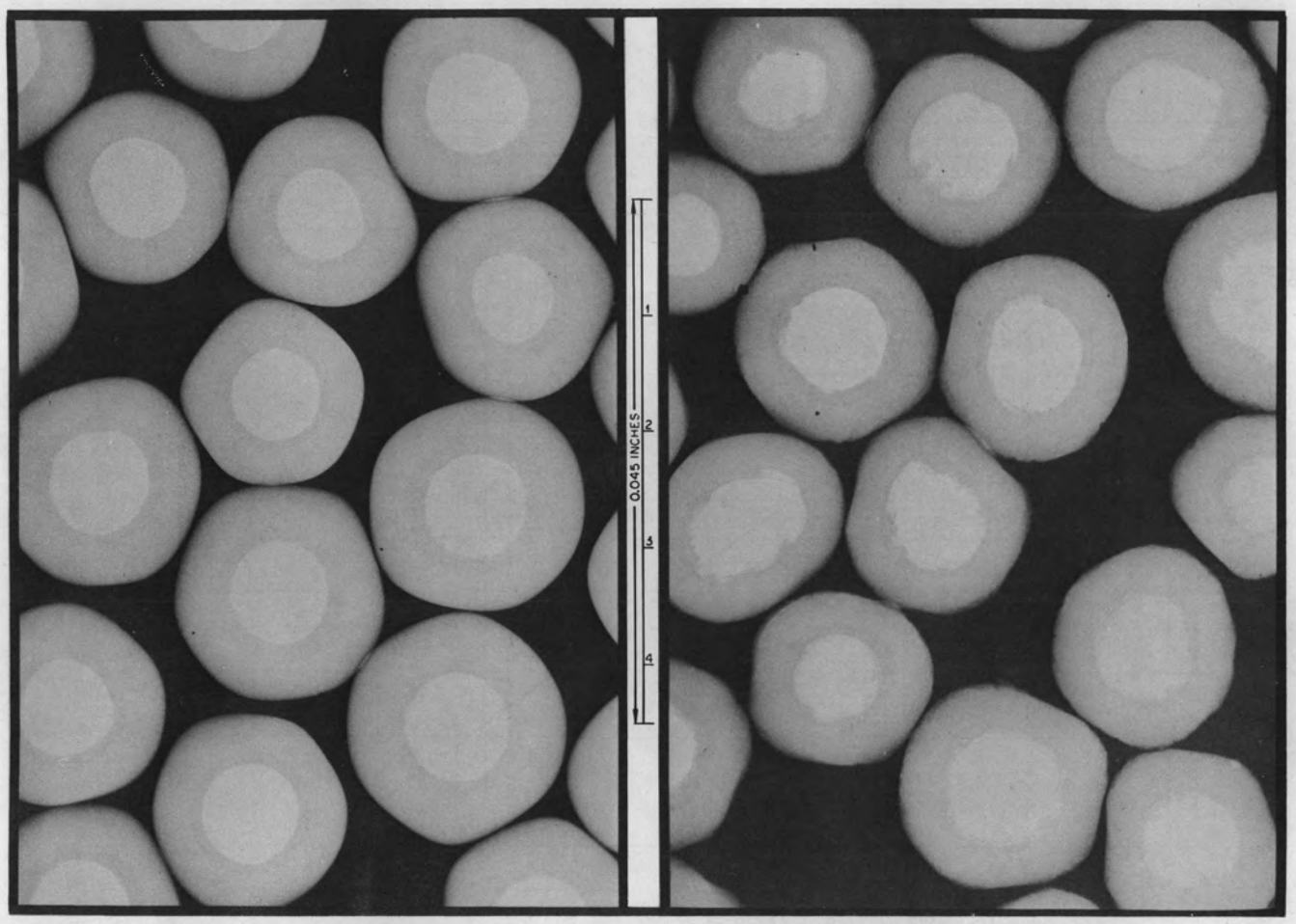


Fig. 7. Relationship Between Fission-Gas Release and Coating-Surface Contamination.

Y-71686



Pyrolytic-Carbon Coated UO_2, Heat Treated 500 hr at 2000°C

Pyrolytic-Carbon Coated UC_2, As-Coated at 2000°C (8 hr)

Fig. 8. Comparison of Fuel Migration at 2000°C in Pyrolytic-Carbon Coated UO_2 and UC_2 Particles.

direct comparison of the performance of carbide and oxide fuel particles with and without thorium as part of the fuel is shown in Fig. 9. In this comparison we have chosen the only available experiment for the thorium-uranium dicarbides which had a similar burnup to the thorium-uranium oxide fuels and also a similar inner coating layer. The greater damage to the coated $(ThU)C_2$ particles must mean that the laminar inner coating on these particles, while similar in appearance and structure, has a lower density. In the case of the mixed-oxide particles, the irradiation experiment in which the coated $(ThU)O_2$ particles were tested also contained coated thoria particles. At the burnup level attained, we were unable to distinguish between the uranium-bearing and the pure thoria particles.

CONCLUSIONS

In summary, then, the irradiation tests on thorium fuels at Oak Ridge National Laboratory closely coupled with process variables and fuel characterizations, indicate that to date:

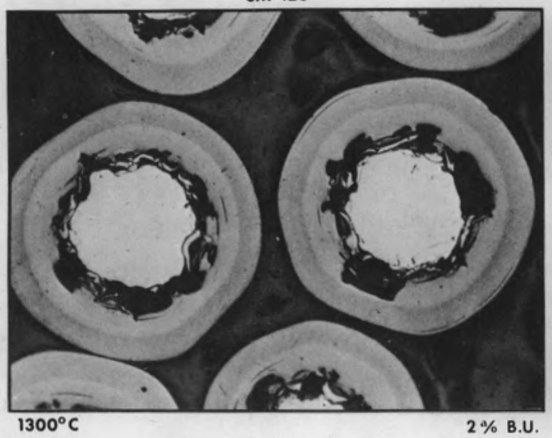
The metal-clad bulk oxide fuel rods using vibratorily compacted sol-gel-derived thoria-base fuels with less than 5% fissile additions will operate at moderate heat ratings to burnups in excess of 100,000 Mwd/tonne (Th+U) with no evidence of breakaway swelling or sudden increases in fission-gas release, and that linear heat ratings in excess of 1000 w/cm can be accommodated without central melting or high fission-gas release rates.

Secondly, the coated-particle irradiations show no significant difference in the performance of fuels with and without thorium. Pyrolytic-carbon-coated oxide fuels perform as well as, and possibly better than, coated dicarbide fuels at burnup levels as high as 25 at. % fissioning.

R-28856

BED-MELTED FUEL

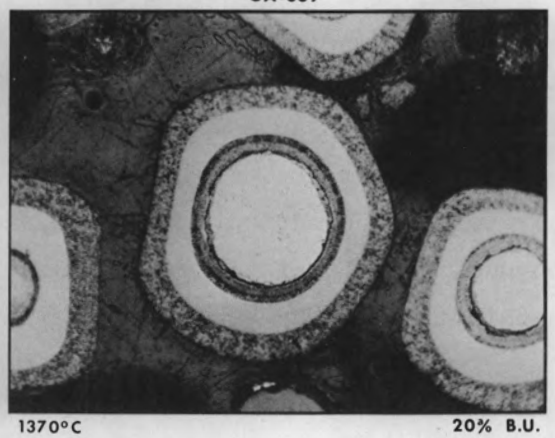
3M-120



1300°C
$(Th,U)C_2$
Th:U = 0.6:1

2% B.U.
HVY. METAL

GA-309

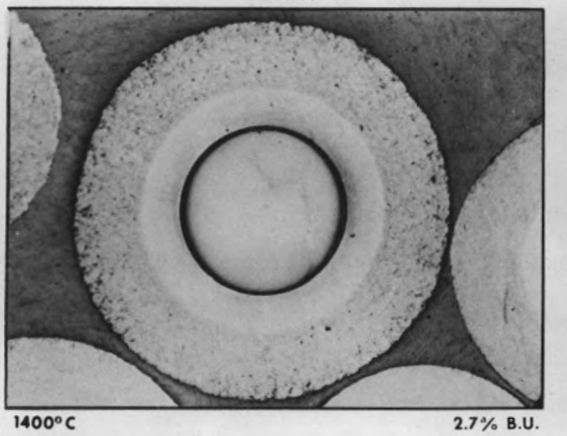


1370°C
UC_2

20% B.U.
HVY. METAL

SOL-GEL FUEL

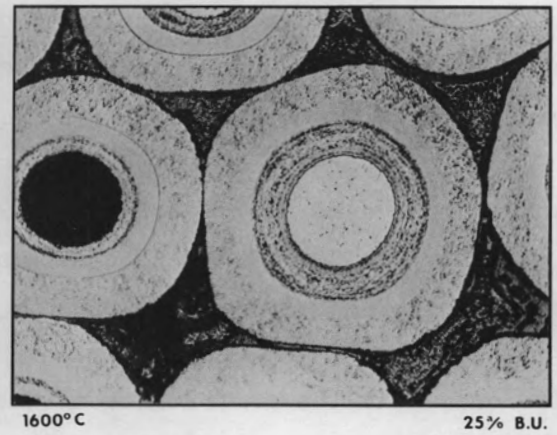
OR-206



1400°C
$(Th,U)O_2$
Th:U = 12:1

2.7% B.U.
HVY. METAL

HB-23



1600°C
UO_2

25% B.U.
HVY. METAL

REFERENCES

1. Rosenthal, M. W., Adams, R. E., Bennett, L. L., Carter, W. L., Douglas, D. A., Jr., Hoskins, R. E., Lawson, C. G., Lotts, A. L., Olson, R. C., Perry, A. M., Roberts, J. T., Salmon, R., and Vondy, D. R., A Comparative Evaluation of Advanced Converters, USAEC Report ORNL-3686, Oak Ridge National Laboratory, January 1965.
2. Rosenthal, M. W., Bauman, H. F., Bennett, L. L., Carlsmith, R. S., and Vondy, D. R., "The Technical and Economic Characteristics of Thorium Reactors," paper presented at the IAEA Panel on "Utilization of Thorium in Power Reactors," Vienna, Austria, June 14–18, 1965.
3. Duret, M. F., and Halsall, M. J., "A Preliminary Assessment of Thorium as a Fuel for Thermal Reactors," paper presented at the IAEA Panel on "Utilization of Thorium in Power Reactors," Vienna, Austria, June 14–18, 1965.
4. Carlsmith, R. S., private communication, February 1966.
5. Peterson, S., Adams, R. E., and Douglas, D. A., Jr., "Properties of Thorium, Its Alloys, and Its Compounds," paper presented at the IAEA Panel on "Utilization of Thorium in Power Reactors," Vienna, Austria, June 14–18, 1965.
6. Wymer, R. G., and Coobs, J. H., "Preparation, Coating, Evaluation, and Irradiation Testing of Sol-Gel Oxide Microspheres," to be published in Journal of the British Ceramic Society.
7. Rabin, S. A., Osborne, M. F., Clinton, S. D., and Ullmann, J. W., "Thorium Fuel Cycle Irradiation Program at the Oak Ridge National Laboratory," Proceedings of the Thorium Fuel Cycle Symposium, Gatlinburg, Tennessee, December 5–7, 1962, USAEC Report TID-7650, Bk. II, p. 643, AEC Technical Information Division.
8. Olsen, A. R., Trauger, D. B., Harms, W. O., Adams, R. E., and Douglas, D. A., Jr., "Irradiation Behavior of Thorium-Uranium Alloys and Compounds," paper presented at the IAEA Panel on "Utilization of Thorium in Power Reactors," Vienna, Austria, June 14–18, 1965.

9. Rabin, S. A., Ullmann, J. W., Long, E. L., Jr., Osborne, M. F., and Goldman, A. E., Irradiation Behavior of High-Burnup ThO_2–4.5% UO_2 Fuel Rods, USAEC Report ORNL-3837, Oak Ridge National Laboratory, October 1965.

10. Osborne, M. F., and Long, E. L., Jr., Postirradiation Examination of Maritime-ORR Loop Experiments 5, 7 and 8, USAEC Report ORNL-TM-921, Oak Ridge National Laboratory, October 1964.

11. Prados, J. W., Beatty, R. L., Beutler, H., Coobs, J. H., Olsen, A. R., and Scott, J. L., "Development of Coated Particle Fuels for Advanced Gas-Cooled Reactors," paper presented at the Second International Thorium Fuel Cycle Symposium, Gatlinburg, Tennessee, May 3–6, 1966.

12. Hamner, R. L., Beatty, R. L., and Beutler, H., "Effects of Heat Treatment on Pyrolytic-Carbon Coated Oxide Particles," pp. 43–50 in GCRP Semiann. Progr. Rept. Sept. 30, 1965, USAEC Report ORNL-3885, Oak Ridge National Laboratory, February 1966.

13. Olsen, A. R., Douglas, D. A., Jr., Hirose, Y., Scott, J. L., and Ullmann, J. W., Properties and Prospects of Thoria-Base Nuclear Fuels, USAEC Report ORNL-TM-1297, Oak Ridge National Laboratory, November 1965.

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